

# METHOD FOR THE PRODUCTION OF A METHANE-CONTAINING GAS

## BACKGROUND OF THE INVENTION

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### 1. Field of the Invention

The present invention relates to the production of a high heating value gas, such as methane gas, from a hydrocarbon-containing feedstock. The feedstock can be selected from a wide range of readily available inexpensive materials such as municipal waste, coal or virtually any other hydrocarbon bearing substance.

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### 2. Description of Related Art

Recently, the United States and other countries have experienced a shortage of natural gas and as a result, natural gas prices for consumers have increased substantially. Accordingly, there is a pressing need for economic methods for the manufacture of a high-value heating gas that can be used in place of natural gas. Natural gas has a composition that includes about 85 percent methane ( $\text{CH}_4$ ), about 10 percent ethane ( $\text{C}_2\text{H}_6$ ) and the balance including propane ( $\text{C}_3\text{H}_8$ ), butane ( $\text{C}_4\text{H}_{10}$ ) and nitrogen ( $\text{N}_2$ ). Methane, the primary component of natural gas, has a heating value of about 51,623 Btu/lb.

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There are many natural resources in addition to natural gas that are utilized to produce energy. For example, coal can be burned in conventional boilers to generate energy from steam. Many regions such as the United States, China and India possess vast reserves of coal that are not presently being exploited for current energy demands due to the high economic and ecological costs related to the use of the coal. Further, some of the coal deposits, particularly those found in India, contain high levels of contaminants and must be subjected to expensive processing prior to utilization.

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There is also an increasing amount of municipal waste being generated by the growing population of the world and the increase in disposable consumer products. The problems and issues relating to the disposal of such waste are well documented. There have been attempts in the prior art to produce useful gas products such as methane from municipal waste products. For example, U.S. Patent No. 3,733,187 by Feldmann discloses a method for converting solid wastes, such as municipal refuse and sewer sludge, to a useful gas product containing methane and ethane. The method involves heating the waste to a temperature of  $500^\circ\text{C}$  to  $670^\circ\text{C}$  and a pressure of 1000 psig to 3000 psig in the presence of a gas containing at least about 90 mole percent hydrogen.

The resulting product gas includes CO, CO<sub>2</sub>, H<sub>2</sub>, H<sub>2</sub>O, CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>. The CO<sub>2</sub> can be scrubbed and the CO can be methanated in the presence of a catalyst. Feldmann discloses some improvements with respect to the foregoing process and apparatus for carrying out the process in U.S. Patent No. 4,005,994 and U.S. Patent No. 4,152,122.

5 U.S. Patent Nos. 5,158,982 and 5,158,983, both by Stapp, disclose a method for treating solid municipal waste, such as scrap automobile tires and polymers, by contacting the waste with hydrogen or hydrogen sulfide at a temperature of 350°C to 450°C to convert the waste to liquid hydrocarbons such as gasoline and diesel range hydrocarbons. It is disclosed that molybdenum can be added to the reaction as a  
10 catalyst.

Thus, it is known that waste products can be treated with a hydrogen-containing gas at high pressures to generate useful product gases and hydrocarbon liquids. Despite the foregoing advances in the art, however, there remains a need for a rapid and economical process for the conversion of municipal waste and other readily available and inexpensive feedstocks to a useful gas product. For example, it would be beneficial if the method were applicable to a variety of feed materials in addition to municipal waste such as coal, including low-grade coals, tar sands, low-grade crude oil and other carbon and hydrocarbon bearing materials that are readily available at low cost or even negative net cost.

## **SUMMARY OF THE INVENTION**

The present invention is directed to a method and apparatus for the conversion of a hydrocarbon bearing feedstock to a useful product gas that includes methane (CH<sub>4</sub>). The feedstock is preferably a low-cost or negative net cost feedstock such as  
25 municipal waste or low-grade coal. The feedstock is contacted with a hydrogen-containing gas, preferably including at least 40 weight percent hydrogen, at an elevated temperature to form a product gas containing CH<sub>4</sub>. The product gas can be used, for example, to generate electricity on-site.

## **DESCRIPTION OF THE DRAWINGS**

Fig. 1 illustrates a method for producing hydrogen gas according to the present invention.

Fig. 2 illustrates a method for producing hydrogen gas according to the present invention.

35 Fig. 3 illustrates a method for producing hydrogen gas according to the present invention.

Fig. 4 illustrates a method for producing hydrogen gas according to the present invention.

Fig. 5 illustrates a method for producing a methane-containing gas product using hydrogen gas according to an embodiment of the present invention.

5 Fig. 6 illustrates a method for producing a methane-containing gas product using hydrogen gas according to an embodiment of the present invention.

Fig. 7 illustrates a method for producing a hydrogen-containing reduction gas stream according to an embodiment of the present invention.

10 Fig. 8 illustrates a method for producing a methane-containing gas product using a hydrogen-containing gas stream according to an embodiment of the present invention.

Fig. 9 illustrates a method for producing a methane-containing gas product from a coal feedstock according to an embodiment of the present invention.

### DESCRIPTION OF THE INVENTION

15 The present invention is directed to the conversion of a hydrocarbon-bearing feedstock to a valuable product gas that includes methane ( $\text{CH}_4$ ). The feedstock is supplied to a hydrogenation unit where it is contacted with large volumes of a gas composition including  $\text{H}_2$  at an elevated temperature such that a portion of the feedstock converts to  $\text{CH}_4$ .

20 One advantage of the present invention is that the feedstock can be virtually any hydrocarbon bearing feedstock, including those that are available at a very low or even a negative net cost.

25 In one embodiment, the feedstock is municipal waste. The municipal waste can include normal household and commercial refuse, hazardous waste, animal waste, sewer sludge, automobile shredder refuse (ASR), scrap rubber tires and the like. Municipal waste is commonly available at a net negative cost since municipalities will typically pay a "tipping" fee for removal and destruction of the waste.

30 According to the United States Environmental Protection Agency, the United States generated 220 million tons of municipal waste in 1998. The amount of waste per person has steadily increased from 2.7 pounds per person per day in 1960, to 3.7 pounds per person per day in 1980 and about 4.5 pounds per person per day in 1998. The components of the municipal waste (before recycling) in the United States in 1998 according to the EPA are listed in Table I.

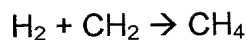
35 Table I Average Municipal Waste Composition

| Component                    | Percentage |
|------------------------------|------------|
| Paper                        | 38.2%      |
| Yard Waste                   | 12.6%      |
| Plastics                     | 10.2%      |
| Food Waste                   | 10.0%      |
| Metals                       | 7.6%       |
| Rubber, Leather and Textiles | 7.0%       |
| Glass                        | 5.7%       |
| Wood                         | 5.4%       |
| Other                        | 3.3%       |

When the feedstock is municipal waste, the feedstock can be treated and separated if necessary. For example, magnetic metals such as iron and steel can be easily removed and sold for scrap. Optionally, the cellulose-based materials (e.g., paper and wood) can also be removed from the feedstock to reduce the level of oxygen compounds in the feedstock. For example, air separation can be used to separate this light, relatively dry fraction of the waste that includes, for example, paper and paperboard, yard trimmings and wood. The remaining material, which includes food wastes, plastics, rubber, leather, non-ferrous metals and glass, can be separated by hydraulic classification. The non-ferrous metals and glass can be recycled off-site.

As is discussed above, the term *municipal solid waste* can include not only residential and general commercial wastes, but specific wastes such as automobile shredder refuse (ASR) or "fluff". ASR, essentially the non-metal components of a recycled automobile, is a heterogeneous mixture of plastics, glass, rubber, fiber, non-recovered metals and dirt. The plastics content of ASR is typically about 20 weight percent and is increasing as the amount of plastic used in automobile construction increases. ASR is presently land-filled and legislation has been proposed in some regions to classify the ASR as a hazardous waste, dramatically increasing the disposal cost. Thus, the conversion of ASR to a useful product gas would enable significant environmental and ecological savings.

The chemical composition of the separated municipal waste feedstock can be generically represented as  $\text{CH}_2$ . Thus, the reaction that occurs when the waste is contacted with large volume of hydrogen gas in the hydrogenation unit can be written as:



Advantageously, a large volume of  $\text{CH}_4$  is produced from the waste and in accordance with the method of the present invention, about 17,000 cubic feet of methane can be generated from 1 ton of municipal waste.

According to another embodiment, the feedstock can be a hydrocarbon bearing substance such as coal, crude oil, tar sand or a similar substance. The coal can be a high-grade or low-grade coal. Coal is abundant throughout the earth's crust and represents a potential low-cost source of energy. However, the use of coal as a basic fuel has been significantly hindered by the environmental problems associated with the burning of coal and the costs associated with removing the contaminants either prior to coal combustion (e.g., treating the coal to remove contaminants) or after combustion (e.g., scrubbing the off-gasses). The method of the present invention enables the conversion of coal, including low-grade coal, to a useful product gas without requiring extensive cleaning of the incoming coal or scrubbing of the combustion off-gasses.

Crude oil is a mixture of liquid hydrocarbons that is extracted from the earth's crust for use as fuel and various petroleum products. Because crude oil is a mixture of widely varying constituents and proportions, the physical properties vary widely and many crude oils have a very low value due to contaminants in the oil. The present invention advantageously enables the treatment of such low-grade crude oils to produce a useful gas product.

Tar sand, also called bituminous sand, is a deposit of loose sand or partially consolidated sandstone that is saturated with highly viscous bitumen. Oil recovered from tar sands is commonly referred to as synthetic crude and is a potentially significant form of fossil fuel. However, the recovery of oil from the tar sand is difficult and cost prohibitive and therefore synthetic oil production from tar sand is not widely practiced. The present invention advantageously enables the production of a useful gas product from otherwise low-value tar sand.

As will be appreciated from the following discussion, a combination of two or more of the above-described components can be used as the feedstock. For example, a separated municipal waste stream can be supplemented with pulverized coal.

One of the important aspects of the method according to the present invention is the in-situ manufacture of large quantities of  $\text{H}_2$  at a relatively low cost. It is believed that one of the primary hindrances to the methods disclosed in the prior art for converting waste materials to useful gas products is the need for high volumes of hydrogen gas and the high cost associated with the hydrogen gas. According to the

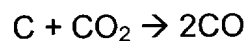
present invention, high volumes of hydrogen gas can be economically generated in-situ. The present invention provides for the formation of a pure H<sub>2</sub> gas stream, or alternatively, a mixture of H<sub>2</sub> and CO.

5 Preferably, a hydrogen gas stream is produced by the reduction of steam (H<sub>2</sub>O) with a metal, thereby forming hydrogen gas (H<sub>2</sub>) and a metal oxide (MeO). In a particularly preferred embodiment, a metal oxide is simultaneously reduced in a separate reactor to form a metal (Me) that can be used for the steam reduction.

10 Referring now to the Figures, the present invention will be described in detail. Figs. 1 and 2 illustrate one embodiment of the method of the present invention using iron and iron oxide to generate hydrogen gas for subsequent hydrogenation of a feedstock. Figs. 1 and 2 illustrate the same apparatus with alternating flow of gaseous reactants and products to continuously form both iron and iron oxide reaction products for the process. Although this embodiment of the present invention is described with respect to oxidation/reduction reactions involving iron metal, it may be desirable to  
15 utilize metal alloys such as Fe/Ni or Fe/Cr.

20 As is illustrated in Fig. 1, steam (gaseous H<sub>2</sub>O) is generated from water 100 in a steam reactor 102, such as a conventional boiler. The steam preferably exits the steam reactor 102 and is conveyed directly to a first reactor 110 through valve 114. The temperature of the steam can be adjusted to control the temperature in the first reactor 110.

25 A reducing gas, preferably a reducing gas that includes carbon monoxide (CO), is generated in a reduction gas reactor 104. In the embodiment illustrated in Fig. 1, supplying carbon 106 and oxygen 108 to the reactor 104 initially generates CO from carbon and oxygen (O<sub>2</sub>). As the reaction progresses, the CO is produced from CO<sub>2</sub> by the Boudouard reaction:

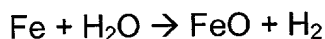


30 The source of the carbon 106 can be, for example, coal, oil, biomass or a similar carbonaceous material. The reducing gas exiting reactor 104 is preferably at least about 90 weight percent CO and more preferably is at least about 95 weight percent CO.

35 The steam and the reducing gas are then supplied to first and second reactors 110 and 112. The first and second reactors 110 and 112 can be adapted to hold a molten metal or molten metal oxide through which reactant gases are passed, or the reactors can be fluidized be reactors adapted to react the gases with particulates in a

fluidized state. For the use of iron and iron oxide, the first reactor **110** and second reactor **112** are preferably fluidized beds.

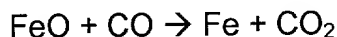
The steam reactor **102** has associated valves **114** and **116** that can be switched to provide the steam to either the first reactor **110** or the second reactor **112**. In the embodiment illustrated in Fig. 1, the valve **114** is open to provide steam to the first reactor **110**. The first reactor is initially provided with iron, preferably in particulate form. In the first reactor **110** the steam reacts with the iron to form iron oxide, preferably FeO, and hydrogen gas, in accordance with the reaction:



In order to maximize hydrogen production, it is preferable to feed a stoichiometric excess of H<sub>2</sub>O to the first reactor **110**. Hydrogen gas and the water vapor are removed through a valve **118** to a condenser **120** where water **122** is removed from the gas stream and pure hydrogen gas **124** is recovered.

Simultaneously, the reduction gas reactor **104** produces CO reducing gas. The valves **126** and **128** are controlled to provide the reducing gas composition to the appropriate reactor. In the embodiment illustrated in Fig. 1, the valve **128** is opened to supply reducing gas to the second reactor **112**. Excess CO **136** is preferably removed to remove oxygen from the system corresponding to the amount of hydrogen being removed from the system. This excess CO **136** can be used as process heat, such as to heat the boiler **102**, advantageously conserving heat value in the process and maximizing the use of unreacted CO.

In the second reactor **112**, iron oxide is initially provided and the reducing gas composition is reacted with the iron oxide to form iron and carbon dioxide, in accordance with the reaction:



The iron oxide is preferably in fluidized particulate form to enable the rapid and economical formation of the iron. Advantageously, the carbon dioxide can be recycled back to the reactor **104** through valve **130** for the production of additional reducing gas. In one embodiment, sufficient CO<sub>2</sub> is recycled back to the reactor **104** such that the amount of fresh oxygen **108** supplied to reducing gas reactor **104** is only enough to maintain the desired reaction temperature. Preferably, the reaction temperature in the reduction gas reactor **104** is from about 800°C to about 1300°C.

Fig. 2 illustrates the identical apparatus as is illustrated in Fig. 1. However, in Fig. 2, the valves **114**, **116**, **118**, **126**, **128**, **130**, **132** and **134** are switched so that the first reactor **110** is the metal reduction reactor and the second reactor **112** is the steam reduction reactor. Hydrogen gas is therefore extracted from the second reactor **112** through valve **134**.

In accordance with the foregoing, it is apparent that the first and second reactors **110** and **112**, at any given point in time during the process, will include some mixture of iron and iron oxide. In one embodiment, the composition of the reactor feed in the first and second reactors is monitored and the flow of gaseous reactants is switched accordingly. Although temperature adjustments to the reactors can be made on a real-time basis, it is an advantage of the present invention that the reactors are maintained at a substantially constant reaction temperature regardless of whether the reactor is being utilized for the reduction of steam or the reduction of iron oxide. Therefore, no heating, cooling and reheating of the non-gaseous reactants is necessary. In a preferred embodiment, the reaction temperature of the first and second reactors is maintained at not greater than about 1000°C, such as from about 700°C to about 900°C.

It is also an advantage of the present invention that the non-gaseous reactants (e.g., the iron and the iron oxide) are not physically moved from one location to another, such as from one reactor or reactor zone to another. On a commercially useful scale, the amount of iron and/or iron oxide in each reactor can be several hundred tons. Eliminating the need to move such a large mass of material substantially reduces the cost associated with producing the hydrogen gas. It will be appreciated that it may be desirable to supplement the iron and/or iron oxide with fresh feed due to any inherent system losses, although such supplementation should be minimal.

Other metal/metal oxide systems can be used for the production of hydrogen gas and subsequent hydrogenation of a feedstock according to the present invention. In one particularly preferred embodiment, tin (Sn) and tin oxide (SnO<sub>2</sub>) is used to form hydrogen gas.

Figs. 3 and 4 illustrate an embodiment of the method of the present invention wherein hydrogen is formed using tin (Sn) and tin oxide (SnO<sub>2</sub>). Similar to Figs. 1 and 2, Figs. 3 and 4 illustrate the same apparatus with alternating flow of gaseous reactants and products to continuously form both metal and metal oxide reaction products.

As is illustrated in Fig. 3, steam is generated from water **300** in a steam reactor **302**, such as a conventional boiler. The steam exits the steam reactor and is conveyed



to the first reactor **310** through valve **314**. The temperature of the steam can be used to partially control the reaction temperature in the first reactor **310**.

A reducing gas, preferably a reducing gas that includes carbon monoxide (CO), is generated in a reactor **304**. In the embodiment illustrated in Fig. 3, supplying carbon **306** and oxygen **308** to the reactor initially generates CO from carbon and oxygen (O<sub>2</sub>). As the reaction progresses, the CO is produced from CO<sub>2</sub> by the Boudouard reaction. The source of the carbon **306** can be, for example, coal or a similar carbonaceous material. The reducing gas exiting the reactor **304** is preferably at least about 90 weight percent CO and more preferably is at least about 95 weight percent CO.

The steam and the reducing gas are then supplied to first and second reactors **310** and **312**. The steam is preferably reacted with molten tin metal by passing the gaseous steam through a pool of the molten tin metal. The molten tin metal will oxidize, forming SnO<sub>2</sub> particulates dispersed in the metal.

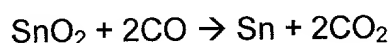
Therefore, the first reactor **310** and second reactor **312** are preferably large reactors adapted to heat and contain molten tin and a mixture (slurry) of molten tin with tin oxide. The steam reactor **302** has associated valves **314** and **316** that can be switched to provide the steam to either the first reactor **310** or the second reactor **312**. In the embodiment illustrated in Fig. 3, the valve **314** is open to provide steam to the first reactor **310**. The first reactor **310** is initially provided with a pool of molten tin having a temperature of at least about 232°C (the melting point of tin) and preferably from about 600°C to about 800°C. In the first reactor **310** the steam reacts with the molten tin to form SnO<sub>2</sub> and hydrogen gas, in accordance with the reaction:



In order to maximize hydrogen production, it is preferable to feed a stoichiometric excess of H<sub>2</sub>O to the first reactor **310**. Hydrogen gas and the excess water vapor are removed through a valve **318** to a condenser **320** where water **322** is removed from the gas stream and pure hydrogen gas **324** is recovered.

Simultaneously, the reduction gas reactor **304** produces CO reducing gas by the Boudouard reaction. The valves **326** and **328** are controlled to provide the reducing gas composition to the appropriate reactor. In the embodiment illustrated in Fig. 3, the valve **328** is opened to supply reducing gas to the second reactor **312**. Excess CO **336** is preferably removed to remove oxygen from the system corresponding to the amount of hydrogen being removed from the system. This excess CO **336** can be used as process heat, such as to heat the boiler **302**. In the second reactor **312**, tin oxide is

initially provided and the reducing gas composition is reacted with the tin oxide to form molten tin metal and carbon dioxide, in accordance with the reaction:



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The tin oxide is preferably in particulate form to enable the rapid and economical reduction of the tin oxide to tin metal. The tin oxide can initially be provided as a slurry of the oxide in molten tin metal. Advantageously, the carbon dioxide can be recycled back to the reactor **304** through valve **330** for the production of additional reducing gas. In one embodiment, sufficient  $\text{CO}_2$  is recycled back to the reactor **304** such that the amount of fresh oxygen **308** supplied to Boudouard reactor **304** is only enough to maintain the desired reaction temperature, which is preferably from about  $800^\circ\text{C}$  to about  $1300^\circ\text{C}$ . This also maximizes the use of unreacted  $\text{CO}$ .

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Fig. 4 illustrates the identical apparatus as is illustrated in Fig. 3. However, in Fig. 4, the valves **314**, **316**, **318**, **326**, **328**, **330**, **332** and **334** are switched so that the first reactor **310** is the metal reduction reactor and the second reactor **312** is the steam reduction reactor. Hydrogen gas is therefore extracted from the second reactor **312** through valve **334**.

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It is apparent that the reactors **310** and **312**, at any given point in time during the process, will include some mixture of tin and tin oxide. In one embodiment, the composition of the reactor feed in the first and second reactors is monitored and the flow of gaseous reactants is switched accordingly. Although temperature adjustments to the reactors can be made on a real-time basis, it is an advantage of the present invention that the reactors are maintained at a substantially constant reaction temperature regardless of whether the reactor is being utilized for the reduction or the oxidation of the tin/tin oxide. Therefore, no heating, cooling and reheating of the non-gaseous reactants is necessary. In a preferred embodiment for the production of hydrogen from  $\text{Sn}/\text{SnO}_2$ , the reaction temperature of the first and second reactors is maintained at a temperature of at least about  $232^\circ\text{C}$  and not greater than about  $1120^\circ\text{C}$  (the melting point of  $\text{SnO}_2$ ), such as from about  $400^\circ\text{C}$  to about  $800^\circ\text{C}$ . Lower reaction temperatures are preferred to minimize large  $\text{SnO}_2$  particles whereas higher temperatures increase reaction kinetics.

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As with the iron/iron oxide system described hereinabove, it is also an advantage of the present invention that the non-gaseous reactants (i.e., the molten tin and the tin oxide) are not physically moved from one location to another, such as from one reactor or reactor zone to another. On a commercially useful scale, the amount of molten tin

and/or tin oxide in each reactor can be several hundred tons. By eliminating the need to move such a large mass of material, the cost associated with producing the hydrogen gas is substantially reduced. It will be appreciated that it may be desirable to supplement the tin and/or tin oxide with fresh feed due to any inherent system losses, although such supplementation should be minimal.

The hydrogen gas stream that is produced according to the foregoing method has a high purity and preferably includes greater than about 99 weight percent hydrogen and more preferably greater than about 99.9 weight percent hydrogen after removal of residual water in the condenser. It is an advantage of this embodiment of the invention that the hydrogen gas does not require separation from another gas species such as carbon monoxide (CO).

The substantially pure hydrogen gas stream that has been formed by reduction of steam can then be used for the conversion of a hydrocarbon containing feedstock to a methane-containing product gas. The integration of the foregoing method and apparatus for the production of hydrogen with a hydrogenation method and apparatus will be more clearly understood with reference to Figs. 5 and 6.

Referring to Fig. 5, water is heated in a boiler **502** to produce steam. The path of the gaseous steam is controlled by valve **514** and in the embodiment illustrated in Fig. 5 is directed to a reactor **510** where a metal (e.g., iron or tin) is oxidized to a metal oxide, thereby forming hydrogen gas in accordance with the method described with respect to Figs. 1-4. The hydrogen gas stream is routed through valve **518** and includes some excess water that can be removed in a condenser **520**. A heat exchange device **532** can be utilized to extract heat value from the mixed hydrogen and steam gas stream, such as to provide heat to the boiler **502**.

Simultaneously, carbon and oxygen are supplied to a reactor **504** where CO is generated and supplied to reactor **512** through valve **526**. In the reactor **512**, a metal oxide is reduced to a metal by reacting with the CO. In one embodiment, such as when the metal is iron, both reactors **510** and **512** are fluidized bed reactors. As is discussed with reference to Figs. 1-4, CO<sub>2</sub> exiting the reactors through valve **516** can be routed to reactor **504** for the production of CO reducing gas by the Boudouard reaction.

The hydrogen gas that is produced by the reduction of steam advantageously has a high purity, and the hydrogen gas stream preferably includes at least about 99 weight percent hydrogen and more preferably at least about 99.9 weight percent hydrogen after removal of excess water.

The hydrogen gas stream is conveyed to a hydrogenation unit **540** where the hydrogen gas is reacted with the incoming feedstock to form methane gas. As is

discussed above, the hydrocarbon-bearing feedstock can be, for example, municipal waste, ASR, scrap rubber tires, coal, oil, tar sand and the like. The type of reactor utilized for hydrogenation will depend upon the nature of the feedstock. Preferably, the hydrogenation unit **540** is a fluidized bed reactor. The hydrogenation unit is preferably  
5 operated at ambient or near ambient pressure, such as a pressure of not greater than about 5 psi. It is an advantage of the present invention that the hydrogenation unit **540** does not have to be operated at elevated pressure in order to produce substantial quantities of a valuable methane-containing product gas.

The reaction in the hydrogenation unit **540** is preferably carried out at a  
10 temperature of at least about 600°C and not greater than about 1100°C and more preferably from about 700°C to about 900°C. The reaction that occurs in the hydrogenation unit is exothermic and therefore the need for additional external heat is minimal. Preferably, at least about 90 weight percent, more preferably at least about 95 weight percent and even more preferably at least about 99 weight percent of the hydrocarbons in the feedstock are converted to methane.

The methane-containing gas stream can be treated in a scrubber **542** to remove  
15 contaminants from the gas stream. For example, the scrubber can apply a caustic solution to form a benign liquid waste stream including, for example, sulfur and chloride compounds. It is advantageous to scrub the methane gas stream prior to combustion, since combustion creates a much higher volume gas stream. Inorganics and other inert  
20 compounds can be removed from the hydrogenation unit **540**. The composition of the residual inorganics and inerts will depend upon the composition of the feedstock and can include carbon, sulfur and ash-forming minerals.

Referring to Fig. 6, the same apparatus as in Fig. 5 is illustrated wherein the  
25 valves **514**, **516**, **518** and **526** are switched such that a hydrogen gas stream is produced by steam reduction in the reactor **512** and is routed to the hydrogenation unit **540**. Thus, a substantially constant flow of a high volume of hydrogen gas can be supplied to hydrogenate the incoming feedstock.

The methane gas formed in the hydrogenation unit can be combusted on-site to  
30 generate electricity, or can be cleaned and provided to end-users as a pipeline gas. Further, a portion of the CH<sub>4</sub> can be recycled back to the other unit operations to provide process heat. For example, a portion of the CH<sub>4</sub> can be diverted to the reactors **510** and **512** and/or can be used to power the boiler **502**.

Figs. 7 and 8 illustrate an alternative embodiment of the present invention  
35 wherein hydrogenation of the feedstock is carried out using a reducing gas that includes a mixture of hydrogen and carbon monoxide (CO).

A flowsheet illustrating the means for generating a H<sub>2</sub>/CO gas stream by the partial oxidation of carbon is illustrated in Fig. 7. Water is provided to a boiler **704** to generate steam. The steam is combined with carbon in a reactor **710** at an elevated temperature, such as from about 900°C to about 1300°C, to form H<sub>2</sub>, CO and excess H<sub>2</sub>O. This gas stream can be passed through a heat exchanger **712** to conserve process heat and then a condenser **714** to remove the excess water. The dried gas stream, which is a mixture of H<sub>2</sub> and CO in a 1:1 molar ratio, can then be used as a hydrogenation gas.

Referring to Fig. 8, a reduction gas stream is provided including H<sub>2</sub> and CO, such as from the partial oxidation of carbon illustrated in Fig. 7. The reduction gas stream is provided directly to the hydrogenation unit **840**. Simultaneously, a feedstock, in this embodiment separated and dried municipal waste, is provided to the hydrogenation unit **840** to be reacted with the reduction gas stream. Preferably, the hydrogenation reaction takes place at a temperature of at least about 600°C and not greater than about 1100°C, such as from about 700°C to about 900°C.

The hydrogenation reaction forms a gas stream that includes CH<sub>4</sub> and CO since the CO essentially passes through the hydrogenation unit **840** without reacting with the feedstock. The gas stream can be scrubbed in scrubber **842** to remove contaminants and form a benign wastewater stream.

In the embodiment illustrated in Fig. 8, the resulting scrubbed product gas stream is split to provide power generation in a combined cycle generator **860** and raise steam in a conventional boiler **862** for power generation via steam turbines. Optionally, a very light waste stream fraction (e.g., cellulose-based materials) can be removed from the municipal waste feedstock and fired in the boiler **862** with the methane. In this embodiment, the treated municipal waste feedstock includes less than about 5 weight percent cellulose-based materials.

The generator **860** can advantageously be a combined cycle generator. In a combined cycle generator, exhaust heat from a first system, referred to as the top cycle, is used to generate power in a second system, referred to as the bottom cycle. Such combined cycle systems typically employ a combustion turbine in the top cycle, and a steam turbine in the bottom cycle. A heat recovery steam generator uses the hot exhaust gas from the combustion turbine to produce steam that drives one or more steam turbines that can be used to generate electricity. Combined cycle generators can operate with up to about 55 percent efficiency, whereas traditional boilers operate at only about 37 percent efficiency.

As is discussed above, the primary feedstock to the hydrogenation unit of the present invention can also be a low-grade hydrocarbon material such as coal, tar sands, low grade crude oil and the like. In the embodiment illustrated in Fig. 8, a backup feedstock stream, such as one that includes pulverized coal and/or crude oil, is provided in the event that there is an interruption in the flow of the primary waste stream due to equipment malfunction or other event.

Fig. 9 illustrates a further embodiment of the present invention for the treatment of coal to form a product gas containing methane. In this embodiment, hydrogen gas is formed by steam reduction/metal oxidation and metal oxide reduction/carbon monoxide oxidation, as is described hereinabove with reference to Figs. 1-4. The coal feedstock **902** is provided to the hydrogenation unit **940** to be contacted with the hydrogen gas. Preferably, the coal feedstock **902** is comminuted such that the maximum particle diameter is not greater than about 1 mm.

Upon treatment of the coal feedstock **902** in hydrogenation unit **940** a methane-containing gas stream is formed. The methane-containing gas stream can be scrubbed in a scrubber **942** to remove gas contaminants. As is illustrated in Fig. 9, a portion of the methane can be recycled to the Boudouard reactor **904** where it is combusted to provide heat for the production of reduction gas.

Also, Fig. 9 illustrates that a portion of the hydrogen gas can optionally be removed before the hydrogenation unit **940**. Hydrogen gas has a high heating value and can be combusted with the methane gas to generate electricity in a generator.

The hydrogenation unit **940** forms a clean (low sulfur) coke product **908** in addition to the methane gas. The clean coke product **908** can be used as a feedstock to the Boudouard reactor **904** where the carbon is converted to carbon monoxide. Further, a portion of the clean coke product **908** can be fired with the methane gas to generate heat for the Boudouard reactor **904**.

Thus, the hydrogenation process of the present invention can produce a clean carbon product, i.e. coke, which has a reduced level of impurities. For example, coal and crude petroleum products frequently include contaminants, such as high levels of sulfur. According to the present invention, the sulfur will react with the hydrogen gas and form hydrogen sulfide that can be easily removed from the gas stream prior to combustion.

When the feedstock is pulverized coal, it is an advantage of the present invention that the hydrogenation treatment of the coal forms a product stream that includes activated carbon. Activated carbon is an amorphous carbon having a porous internal structure and a high surface area. Activated carbon has a high value and can be used

for air and water purification, waste treatment, removal of mercury (Hg) and SO<sub>x</sub> from stack gases and the like. Thus, the hydrogenation process when applied to coal provides a high Btu value gas stream as well as a valuable by-product.

- 5 While various embodiments of the present invention have been described in detail, it is apparent that modifications and adaptations of those embodiments will occur to those skilled in the art. However, it is to be expressly understood that such modifications and adaptations are within the spirit and scope of the present invention.